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Spin-dependent dynamics of ultrafast polarised optical pulse propagation in coherent semiconductor quantum systems

G. Slavcheva* and O. Hess

Advanced Technology Institute, School of Electronics and Physical Sciences, University of Surrey,
Guildford GU2 7XH, Surrey, United Kingdom

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A new model for rigorous theoretical description of circularly (elliptically) polarised ultrashort optical pulse interactions with the resonant nonlinearities in semiconductor optical waveguides is proposed. The method is based on self-consistent solution in the time domain of the vector Maxwell equations coupled via microscopic polarisation to the coherent time-evolution equations of a discrete N-level quantum system in terms of the real pseudospin (coherence) vector. The model is initially applied to a generic two-level quantum system and subsequently to a 4-level system describing the heavy-hole excitonic transitions in low-dimensional semiconductor systems, such as quantum wells and quantum dots. Selective optical excitation of specific spin states by predefined helicity of the optical field in the linear regime and an onset of self-induced transparency and polarised soliton formation in the nonlinear regime are numerically demonstrated in both discrete-level systems.

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1 Introduction

The all-optical spin orientation using circularly (elliptically) polarised optical pulses is a technique of key importance for generation and manipulation of spin-polarised states in semiconductor nanostructures in view of achieving quantum coherent control. A necessary condition for quantum coherence is the use of sufficiently short optical pulses so they can interact with the quantum system before it can be affected by its environment. Coherent-carrier control in semiconductor quantum dots (QDs) has recently attracted significant interest because it allows coherent manipulation of the carrier wave functions on a time scale shorter than typical dephasing times. This in turn is a prerequisite for successful implementation of ultrafast optical switching and quantum-information processing.

The dipole optical selection rules for the excitonic transitions in bulk semiconductors, and in particular in low-dimensional semiconductor systems, require total angular-momentum projection difference along the quantization axis (coinciding with the optical field propagation direction) $\Delta J_z = \pm 1$. These transitions are excited by circularly polarised light and can no longer be described by a single scalar interaction (valid for the linear polarisation case). Therefore a new model of the coherent dynamics is needed to account for the polarisation state of the applied electromagnetic field. We propose and develop a new model for rigorous theoretical description of circularly (elliptically) polarised ultrashort optical pulse interactions with the resonant nonlinearities in planar optical waveguides and semiconductor microcavities. The method is based on the self-consistent Finite-Difference Time-Domain (FDTD) [1] solution of the vector Maxwell equations coupled via macroscopic polarisation to the time-evolution equations of a discrete N-level quantum system [2, 3] in terms of the real pseudospin (coherence) vector [4]. It takes

* Corresponding author: e-mail: G.Slavcheva@surrey.ac.uk, Phone: +44 1483 686086, Fax: +44 1483 9404

properly into account the vector nature of the electromagnetic field and hence the polarisation state of the optical excitation.

2 Theoretical model

Consider a plane electromagnetic wave propagating along z direction and elliptically polarised in a plane perpendicular to z . The optical wave carrier frequency ω_0 is tuned in resonance with the fundamental

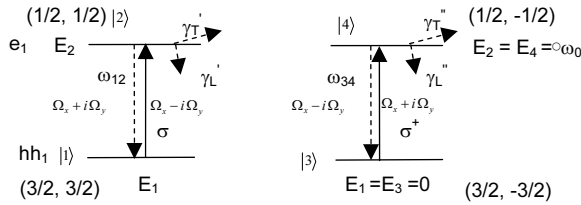


Fig. 1 Energy-level diagram of σ^- and σ^+ heavy-hole excitonic transitions in a quantum well or quantum dot. The levels are labeled by pairs of numbers representing the total angular momentum and its projection along the propagation z -axis. The dipole-allowed transitions correspond to $\Delta J_z = \pm 1$ and the energy separation is $\hbar\omega_0$. The coherent optical transitions excited by left (σ^-)- and right (σ^+)-circularly polarised light and the relaxation processes are designated by arrows (Ω_x and Ω_y – real and imaginary part of the Rabi frequencies associated with the transitions). The dissipation in the system is accounted for through the population relaxation (longitudinal) time γ_L and the transverse (dephasing) time γ_T .

heavy-hole excitonic transition of the semiconductor system. The band structure in the vicinity of the centre of the Brillouin zone ($\mathbf{k}=0$) is isomorphic with a pair of two-level systems corresponding to σ^+ and σ^- -heavy-hole excitonic transitions (Fig. 1). For a pulse tuned with the heavy-hole excitonic resonance in a typical 100 Å GaAs quantum well, the energy interval is $\Delta E = E_2 - E_1 = 1.475$ eV, the corresponding wavelength is at $\lambda \approx 842.5$ nm, which yields a resonant pulse centre frequency $\omega_0 = 2.24 \times 10^{15}$ rad.s $^{-1}$.

The resonant nonlinearity is modelled by an ensemble of multi-level systems (4-level systems in this particular case) with density N_a . The time

evolution of a discrete N -level system in an external perturbation is governed by the equation of motion, derived in [4] employing the generalised pseudospin formalism based on the commutator Lie algebra of $SU(N)$ group. In the case of a four-level system, the dynamics is described in terms of the $SU(4)$ group yielding the following equation of motion for the 15-dimensional real state (coherence) vector:

$$\frac{\partial S_i}{\partial t} = f_{ijk} \gamma_j S_k - \frac{1}{T_i} (S_i - S_{ie}) \quad i, j, k = 1, \dots, 15 \quad (1)$$

where the dissipation in the system is accounted for by introducing non-uniform relaxation times T_i as phenomenological parameters, describing the relaxation of the real vector components toward their equilibrium values S_{ie} and f is the fully antisymmetric tensor of the structure constants of $SU(4)$ group. The pseudospin vector \mathbf{S} and the torque vector $\boldsymbol{\gamma}$ are expressed in terms of the λ -generators of the $SU(4)$ Lie algebra.

Assuming that the initial population resides on the lower-lying levels, the system Hamiltonian is given by:

$$\hat{H} = \hbar \begin{pmatrix} 0 & -\frac{1}{2}(\Omega_x - i\Omega_y) & 0 & 0 \\ -\frac{1}{2}(\Omega_x + i\Omega_y) & \omega_0 & 0 & 0 \\ 0 & 0 & 0 & -\frac{1}{2}(\Omega_x + i\Omega_y) \\ 0 & 0 & -\frac{1}{2}(\Omega_x - i\Omega_y) & \omega_0 \end{pmatrix} \quad (2)$$

where $\Omega_x = \frac{\wp}{\hbar} E_x$; $\Omega_y = \frac{\wp}{\hbar} E_y$ and \wp is the exciton dipole transition matrix element. Using the above Hamiltonian the torque vector is calculated and substituted back in (1), taking into account all non-vanishing components of the f -tensor. Thus a system describing the time-evolution of the 15-dimensional state vector is obtained. For the special case of a two-level system the Hamiltonian is 2×2 matrix, the λ -generators of the SU(2) Lie algebra are simply the Pauli matrices and the pseudospin system is given in [5]. Vector Maxwell equations for the optical wave propagation [2] coupled via polarisation to the pseudospin equations (1) are solved self-consistently in the time domain employing the FDTD technique [1].

3 Simulation results for optically induced spin dynamics

The initial boundary value problem requires the knowledge of the whole time history of the initial field along some characteristic, e.g. at $z = 0$. The circularly-polarised optical pulse is modelled by two orthogonal linearly polarised optical waves, phase-shifted by $\pi/2$:

$$\sigma^+ \begin{cases} E_x(z=0, t) = E_0 \text{sech}(10\Gamma) \cos(\omega_0 t) \\ E_y(z=0, t) = E_0 \text{sech}(10\Gamma) \sin(\omega_0 t) \end{cases}$$

$$\sigma^- \begin{cases} E_x(z=0, t) = E_0 \text{sech}(10\Gamma) \cos(\omega_0 t) \\ E_y(z=0, t) = -E_0 \text{sech}(10\Gamma) \sin(\omega_0 t) \end{cases} \quad (3)$$

where E_0 is the initial field amplitude, $\Gamma = [t - (T_p/2)] / (T_p/2)$ and T_p is the pulse duration. The simulation domain is $150 \mu\text{m}$ long with resonantly absorbing/amplifying two-level or four-level medium embedded between two free-space regions each with length $7.5 \mu\text{m}$. The carrier frequency resonant with the heavy-exciton dipole transition is taken to be $\omega_0 = 1.2558 \times 10^{15} \text{ rad.s}^{-1}$, corresponding to a wavelength $\lambda = 1.5 \mu\text{m}$.

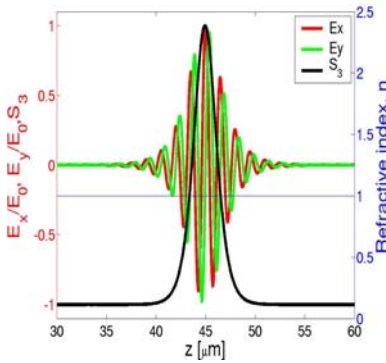


Fig. 2 A snapshot of a polarised soliton at $t = 200 \text{ fs}$ propagating in absorbing medium. The 2π SIT left-circularly polarised pulse completely excites and de-excites the two-level system of quantum absorbers thereby performing a full Rabi flop of the population inversion.

3.1 Two-level system

We have numerically demonstrated selective excitation of the dipole transitions with $\Delta J_z = 1$ or $\Delta J_z = -1$ by predefined helicity of the optical pulse in the linear regime [2]. In the nonlinear regime, when the self-induced transparency (SIT) condition is satisfied, namely the pulse duration $T_p \ll T_1, T_2$ ($T_p = 100 \text{ fs}$, $T_1 = T_2 = 100 \text{ ps}$). Choosing the pulse amplitude according to the Pulse Area Theorem to give a 2π pulse area, which yields $E_0 = 4.2186 \times 10^9 \text{ Vm}^{-1}$, we have numerically demonstrated formation of polarised SIT-solitons (Fig. 2).

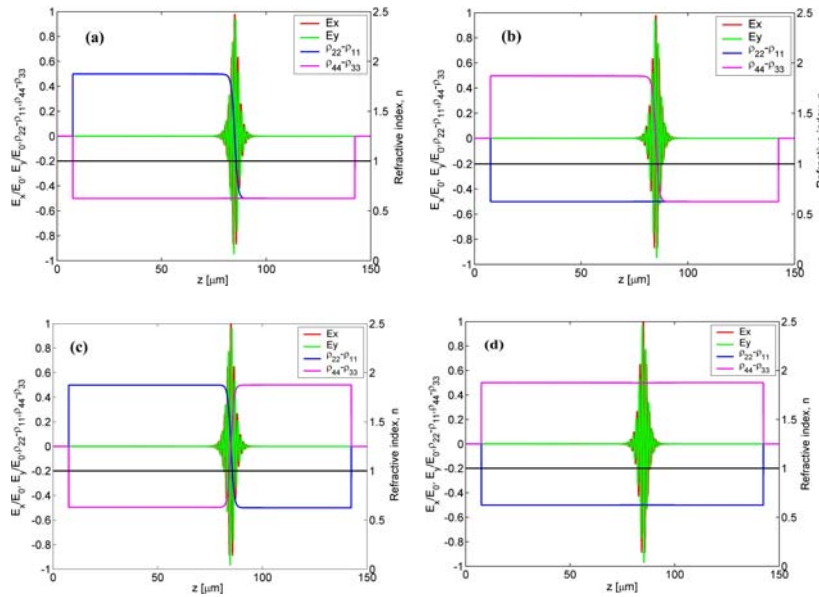


Fig. 3 A snapshot of a circularly polarised pulse at the simulation time $t = 333$ fs. The E-field components are plotted together with the population inversion $\rho_{22} - \rho_{11}$ in the first and $\rho_{44} - \rho_{33}$ in the second two-level systems, indicating the boundaries of the active medium ($n = 1$). (see the text for discussion).

3.2 Degenerate four-level system in the nonlinear regime

We investigate the high-intensity nonlinear regime of pulse propagation. The SIT criterion is satisfied using an ultrashort pulse with $T_p = 100$ fs and setting the relaxation times $T_{13} = T_{14} = T_{15} = 100$ ps, $T_1 = T_6 = T_7 = T_{12} = 100$ ps, while setting the rest (describing crossed or spin-flip transitions) to infinity, thereby reducing the system to a pair of two independent two-level systems. We choose the initial pulse amplitude to correspond to a π -pulse according to the Pulse Area Theorem, giving $E_0 = 1.093 \times 10^9$ Vm $^{-1}$. A π -pulse excites (or de-excites) completely the two-level system [6]. The simulation results for the ultrashort pulse propagation are shown in Fig. 3, for both polarisations of the injected ultrashort pulse σ^- and σ^+ . In Fig. 3(a, b) all the population is assumed initially in the ground states. In Fig. 3(a) a left-circularly polarised pulse σ^- (Eq. (3)) is injected. The pulse excites the first two-level system bringing the population to the upper state and does not affect the second system (see Fig. 1). Figure 3(b) shows a right-circular pulse (σ^+ in Eq. (3)) injected into the same system, the first two-level system is not affected, while the population of the second system is driven into the upper level. In Fig. 3 (c, d) the initial population is assumed equally distributed between level $|1\rangle$ and level $|4\rangle$. Figure 3(c) depicts a σ^- -pulse exciting the first two-level system into the upper level and de-exciting the second one into the ground state. Figure 3(d) shows the failure of a σ^+ -pulse to affect the population in both systems. The remaining two cases, namely: population equally distributed between level $|2\rangle$ and level $|3\rangle$ and all population distributed equally

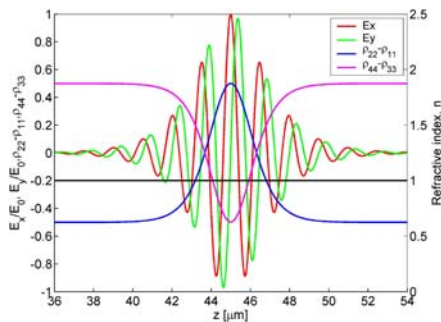


Fig. 4 A magnified snapshot of a 2π left-circularly polarised σ^- pulse propagating in a 4-level medium with initial population $\rho_{11e} = 1/2$, $\rho_{44e} = 1/2$ at the simulation time $t = 200$ fs.

population is assumed equally distributed between level $|1\rangle$ and level $|4\rangle$. Figure 3(c) depicts a σ^- -pulse exciting the first two-level system into the upper level and de-exciting the second one into the ground state. Figure 3(d) shows the failure of a σ^+ -pulse to affect the population in both systems. The remaining two cases, namely: population equally distributed between level $|2\rangle$ and level $|3\rangle$ and all population distributed equally

between the upper-lying levels are considered analogously. The results show that specific spin states can be excited by choosing the proper polarisation of the injected ultrashort optical pulse. Thus we can coherently control the spin population of specific states in the four-level quantum system.

We have numerically demonstrated self-induced transparency in a four-level system, initially prepared in a state with equally distributed population between the lower (upper)-lying level in the first two-level system and the upper (lower)-lying level in the second system (Fig. 4). The ultrashort polarised pulse travels unchanged in the degenerate four-level medium driving locally the population through full Rabi-flops in both two-level systems.

4 Conclusion

We have developed a theoretical model of ultrafast polarised optical pulse interactions with multi-level quantum systems employing the generalised pseudospin formalism [4]. Coherent vector Maxwell-pseudospin equations have been derived and solved in the time domain for the case of a degenerate four-level system used to model the excitonic σ^- and σ^+ -transitions in semiconductor structures. We have numerically demonstrated selective excitation of specific spin-states with predefined helicity of the optical pulse. In the nonlinear regime, we have demonstrated formation of polarised SIT-solitons in a two-level system and in a specially prepared degenerate four-level system. The proposed formalism can be easily extended to more spatial dimensions, more complex discrete-level configurations and structure geometries. The validity of the method spans to the extreme nonlinear regime. The model fully accounts for the polarisation state of the optical excitation and allows modelling of the non-equilibrium dynamics associated with coherent optical spin generation and relaxation.

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